

# Biogas as a reproducible energy source: Its steam reforming for electricity generation and for farm machine fuel

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**Abstract.** The use of biogas as an energy source reduces the chance of possible emission of two greenhouse gases, CH<sub>4</sub> and CO<sub>2</sub>, into the atmosphere at the same time. Its nature of being a reproducible energy source makes its use even more attractive. There exist two ways of using biogas as an energy source: (1) as a natural gas substitute, to burn it in a boiler or in a stove to obtain heat or to supply it to a gas engine for power generation, and (2) to steam-reform it, to obtain hydrogen for fuel cell power generation or to obtain syngas (CO + H<sub>2</sub>) from which dimethylether is synthesized to be used as diesel fuel substitute for farm machines. The former way of utilization has been commonly employed worldwide. In this article, we present the latter way that involves steam reforming of biogas for its further use as an energy source. Necessary processes involved for this way of utilization are outlined, from steam reforming of biogas to the production of hydrogen or dimethylether. The chemical uniqueness of the biogas as a feedstock for steam reforming was identified, along with the difficulties to be overcome for its practical implementation. © 2006 Published by Elsevier B.V.

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## 1. Introduction

Steam reforming of methane (and other carbon sources) to produce hydrogen or syngas (a mixture of CO and H<sub>2</sub>) is a well-established technology [1]. An enormous amount of hydrogen or syngas for industrial use is produced through this route from natural gas, naphtha or coal. Biogas is generally a mixture of methane (CH<sub>4</sub>) and carbon dioxide (CO<sub>2</sub>), similar in

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Table 1  
Compositions of biogas from various biogas plants

Component	Composition (%)		
	Obihiro University of Agriculture and Veterinary Medicine <sup>a</sup>	Piggery in Gunma Prefecture <sup>b</sup>	Joetsu City Sludge Recycle Park <sup>c</sup>
H <sub>2</sub>	Not detected	Not detected	0.14
CH <sub>4</sub>	60.2	66.9	61.4
CO <sub>2</sub>	39.8	33.1	38.5

<sup>a</sup> Cow manure, sampled on January 7, 2005.

<sup>b</sup> Pig manure, sampled on February 15, 2005.

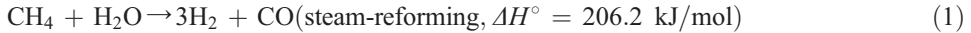
<sup>c</sup> Sludge and wet refuse, sampled on February 15, 2005.

composition to natural gas, and thus should be steam-reformed easily without major modifications of the existing technology.

The merit of steam-reforming biogas, instead of simply using it as a natural gas substitute, lies in its additional freedom to be employed as a renewable energy source. Produced hydrogen can be used to drive fuel cells, through which electricity and hot water can be produced at higher efficiency than by gas engines or microturbines. Syngas can be converted into dimethylether that could be used as diesel fuel substitute in farm machines. Thus, a self-supporting farm may be realized this way, in terms of its energy consumption.

## 2. Characteristics of steam reforming reaction

Steam reforming of methane involves the following reactions:



Eq. (1) produces syngas and Eq. (2) converts CO to CO<sub>2</sub> in order to further squeeze hydrogen from water. While CO<sub>2</sub> is also an important green house gas, this conversion (1 mol of CO<sub>2</sub> from 1 mol of CH<sub>4</sub>) is favourable as CH<sub>4</sub> has the higher global warming potential than CO<sub>2</sub>. If H<sub>2</sub> is the objective, Eqs. (1) and (2) together will produce 4 mol of H<sub>2</sub> from 1 mol of CH<sub>4</sub>.

## 3. Steam-reforming of biogas

Steam-reforming of biogas was examined using a CH<sub>4</sub>/CO<sub>2</sub> mixture that simulates a typical biogas composition. A few examples of biogas compositions are listed in Table 1. The steam-reformed biogas in this study simulates the one that is obtained from the biogas model plant installed at the Field Science Center, Obihiro University of Agriculture and Veterinary Medicine [2]. The plant processes manure from about 70 cows.

Table 2  
Compositions of steam reforming products from the simulated biogas

Component	Composition (%)			
	600 °C		800 °C	
	4000 h <sup>-1</sup>	9000 h <sup>-1</sup>	4000 h <sup>-1</sup>	9000 h <sup>-1</sup>
H <sub>2</sub>	59.6	55.1	63.6	64.2
CO <sub>2</sub>	24.4	26.2	16.9	15.6
CO	10.0	7.4	19.5	20.1
CH <sub>4</sub>	6.0	11.3	0.0	0.1

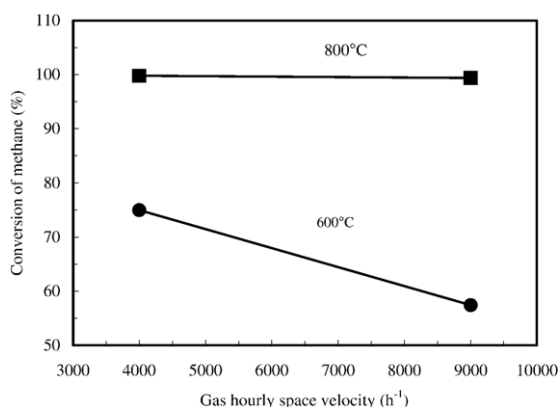


Fig. 1. Methane conversion in a steam reforming of the simulated biogas.

Table 2 and Fig. 1 show the results of the simulated-gas steam reforming performed at two temperatures as a function of gas-hourly space velocity (GHSV) with the steam to methane ratio of 3. Nickel-on- $\alpha$ -alumina (Sud Chemie AG, FCR-4-02) is used as a catalyst. Steam reforming over this type of catalyst is very fast, and at the GHSV of  $4000 \text{ h}^{-1}$  it almost reaches equilibrium. It is found in Fig. 1 and Table 2 that at the temperature of  $800^\circ\text{C}$  almost all the methane is converted to CO, giving off three times as much hydrogen to be used for fuel cell power generation. Note that at  $800^\circ\text{C}$ , Eq. (1) is dominant, with very small amount of water-gas shift reaction occurring, as may be found from the  $\text{H}_2/\text{CO}$  ratio being close to 3 as expected from Eq. (1).

#### 4. Dimethylether synthesis from syngas

Once syngas ( $\text{CO} + \text{H}_2$ ) is obtained by the steam reforming of biogas, dimethylether ( $\text{CH}_3\text{OCH}_3$ , DME) can be synthesized either through a one-step or a two-step process.



Dimethylether possesses characteristics similar to diesel fuel, as may be found in Table 3. Its cetane number is almost the same as, or even more favorable than, that of diesel fuel. Furthermore, DME does not contain sulfur (see later) and therefore does not emit  $\text{SO}_x$  when burned in diesel engines. It also gives almost no particulates, since it does not contain polyaromatics that produces soot.

Table 3  
Characteristics of dimethylether and diesel fuel

Properties	Dimethylether	Diesel fuel
Boiling point ( $^\circ\text{C}$ )	-25.1	180–370
Specific gravity (liquid, $\text{g}/\text{cm}^3$ , at $20^\circ\text{C}$ )	0.67	0.84
Explosion limit (%)	3.4–17	0.6–6.5
Cetane number	55–60	40–55
Lower heating value ( $\text{MJ}/\text{kg}$ )	28.90	41.86

Table 4  
Sulfur content in the biogas obtained from various plants before and after desulfurization

Sulfur type	Content (ppm)					
	Obihiro University of Agriculture and Veterinary Medicine <sup>a</sup>		Piggery in Gunma Prefecture <sup>b</sup>		Joetsu City Sludge Recycle Park <sup>b</sup>	
	Before	After	Before	After	Before	After
Inorganic						
H <sub>2</sub> S	1531	24.6	4011	5.4	99.6	3.5
Organic						
CH <sub>3</sub> SH	11.66	0.50	0.92	Not detected	0.70	0.01
C <sub>2</sub> H <sub>5</sub> SH	0.99	0.36	0.40	0.07	0.33	0.17
Others <sup>c</sup>	0.35	0.33	1.28	0.40	2.61	0.18

<sup>a</sup> Biotidesulfurization.

<sup>b</sup> Dry desulfurization.

<sup>c</sup> Approximate value.

Major differences between DME and diesel fuel are their boiling point and chemical structure. At normal temperature and pressure, DME is a gas, and in order to liquefy it has to be cooled down to below  $-25.1^{\circ}\text{C}$  or pressurized at 0.62 MPa. This will obviously necessitate the modification of fuel tanks and pipes. Furthermore, different chemical nature of DME from that of common diesel fuel may also necessitate the use of proper materials for the engine parts that are in contact with the fuel.

## 5. Problems associated with the steam reforming of biogas

One of the major obstacles for steam reforming of biogas is its high sulfur content. Table 4 lists inorganic (H<sub>2</sub>S) and organic sulfur contents in the biogas listed in Table 1. The biogas plant at Obihiro is equipped with a biotidesulfurization unit, while the other two are equipped with dry desulfurizer that employs iron-based adsorbent. It is apparent from Table 4 that H<sub>2</sub>S contents in the biogas from animal manure are very high before desulfurization. After desulfurization, it is reduced to the level of 10 ppm. Biotidesulfurization and dry desulfurization appear to be comparable in the effectiveness of desulfurization, although the latter may incur much higher running cost than the former technique.

Noteworthy is the presence of organic sulfur compounds in the biogas. Six organic sulfur species were observed, including mercaptans and sulfides. It is further noted in Table 4 that organic sulfurs were less easily removed as H<sub>2</sub>S was, and heavier sulfur compounds are less well removed either by biotidesulfurization or by dry desulfurization process.

The above sulfur level after desulfurization is comparable or even better than common natural gas or petroleum-based fuels. At this level, steam reforming of biogas should be a fairly simple and easy task, leading to power generation through fuel cells or to the production of DME.

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